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Kinetics of photo-stimulated adsorption of enzyme molecules onto *n*- and *p*-type silicon

A.V. Kozlowski, S.V. Stetsyura

Saratov State University, 410012, Saratov, Russia
kozlowsky@bk.ru

Atomic force microscopy (AFM) is an effective method for monitoring the deposition of functional organic layers on a silicon substrate [1]. Enzyme molecules (for example, glucose oxidase (GOx)) can be used to fabricate a receptor layer on the surface of an ion-sensitive semiconductor transducer. The adsorption of enzyme molecules onto the semiconductor surface is due to electrostatic interaction. Thus, the conductivity type of a semiconductor substrate, as well as a change in surface charge density induced by illumination, can influence the number of adsorbed enzyme molecules.

In our work, AFM was used to study the kinetics of photo-stimulated adsorption (PSA) of GOx molecules onto the surface of both *p*- and *n*-type silicon.

The experiments were performed with single-crystal silicon wafers of *n*-type (ρ = 4 Ω cm) and *p*-type (ρ = 8 Ω·cm). Initially, the substrates were boiled in a peroxide–ammonia solution and rinsed in deionized water (resistivity 18.2 MΩ·cm). This treatment leads to "reconstruction" of a native oxide layer while the silicon surface acquires negative charge in deionized water due to activation of OH-groups. GOx molecules from *Aspergillus niger* was used as enzyme molecules. In a wide pH range of the solution, the GOx molecule has an effective negative charge. The size of the GOx molecule is 6.0×5.2×7.7 nm^3 [2]. A cationic polyelectrolyte polyethylenimine (PEI) with a molecular weight of 25 kDa was used to increase the adsorption of negatively charged GOx onto silicon substrates. The PEI molecules were adsorbed on silicon substrates from the 1 mg/ml aqueous solution during 10 min followed by rinsing in water during 10 min and drying. The photo-assisted layer-by-layer adsorption technique suggested in [3] was used to adsorb GOx from the 0.5 mg/ml aqueous solution onto covered with PEI silicon substrates. The adsorption time varied between 10 min and 60 min.

The topography of the films was measured using AFM by NTEGRA Spectra (NT-MDT Spectrum Instruments, Russia). Scanning was performed under ambient conditions at a frequency of 0.5 Hz in taping mode using HA_NC/W_2C cantilevers of ETALON series. The Gwyddion software for statistical analysis of AFM data was used.

According to the AFM images, the surface coverage of substrate by GOx molecules taking into account the GOx size and the limitations of the measurement method was determined. The surface coverage with GOx molecules calculated as a percentage of area covered with GOx from the total substrate area. It was obtained that, for the p-Si type, the surface coverage value decreases, while for n-Si, it increases with adsorption time at fixed illumination level.

Thus, using the method of scanning probe microscopy, the quantitative results of the GOx adsorption process as a function of illumination, adsorption time, and the conductivity type of Si substrate were obtained.

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